Transition Metals play central roles in numerous biological processes, participating in catalysis at active sites of metalloenzymes as well as engaging in cellular signaling events. Of note, the enzymatic C–H hydroxylation reactivity of cytochrome P450 has inspired the development of strategies for the selective and efficient conversion of hydrocarbon feedstocks into value-added products. To this end, our investigations of high-spin iron dipyrrinato complexes revealed the importance of a unique high-spin ferric iminyl electronic configuration for C–H amination processes. Studies also highlighted the access to a di-iron bridging imido that can catalytically transfer the N-group into allylic and benzylic C–H bonds. Understanding the electronic structure considerations in our systems permitted us to tune the iron dipyrrin complexes to accomplish a diastereoselective C–H amination protocol. Beyond transition metal catalysis, motivated by the implications of copper ions in diseases, we sought to elucidate the emerging role of copper in interacting with proteins and modulating enzymatic activity. To this end, we explored activity-based protein profiling approaches to identify Cu-regulated metalloproteins and proposed methods to leverage the Cu-dependency of such targets for therapeutics.